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(54) Cavity-locked ring down spectroscopy

(57) A cavity ring-down spectroscopy system comprising a cavity for holding a sample, a light source for generating locking light and sampling light of different polarization and/or of different frequencies, locking

means for locking the cavity and the light source, and detection means for detecting sampling light and for allowing determination of ring-down rates indicative of an absorption of the sampling light by the sample.

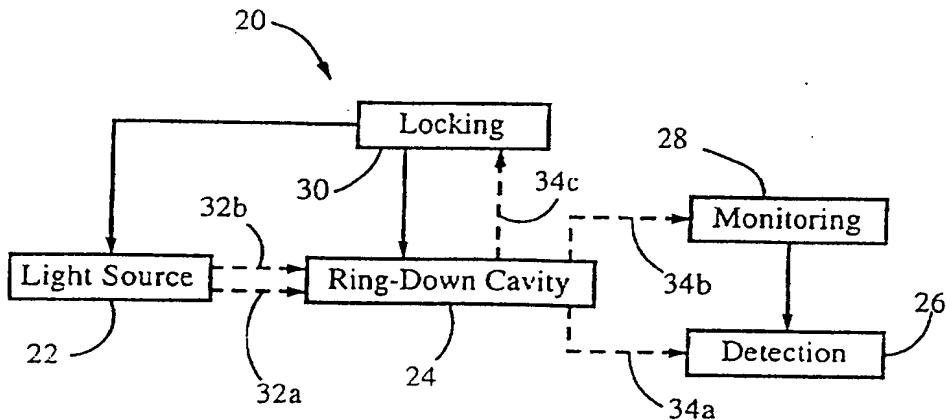


FIG. 1-A

Description

[0001] This invention relates to cavity ring-down spectroscopy (CRDS), and in particular to systems and methods for locking a laser and a ring-down cavity for CRDS.

[0002] Traditional spectroscopic methods are limited in sensitivity to approximately one part per ten thousand (1:10⁴) to one part per hundred thousand (1:10⁵), whereas cavity ring-down spectroscopy (CRDS) allows making absorption measurements with sensitivities on the order of one part per ten million (1:10⁷) to one part per billion (1:10⁹) or higher.

[0003] In a conventional CRDS system, the sample (absorbing material) is placed in a high-finesse stable linear optical resonator. The intensity of a light pulse introduced in the resonator decreases in time. For an empty cavity, the intracavity light intensity follows an exponential decay characterised by a ring-down rate that depends only on the reflectivity of the mirrors, the separation between the mirrors, and the speed of light in the cavity. If a sample is placed in the resonator, the ring-down is accelerated. Under suitable conditions, the light decay remains exponential. An absorption spectrum for the sample is obtained by plotting the reciprocal of the ring-down rate versus the wavelength of the incident light.

[0004] Conventional pulsed CRDS systems provide a number of challenges. In systems using pulsed lasers, data acquisition rates may be limited by the repetition rate of the pulse laser source. Moreover, the intensity of the light coupled into and out of the cavity may be small, as a consequence of the relatively low spectral overlap between cavity modes and laser linewidth, as well as a lack of significant light buildup within the cavity.

[0005] US Patent No. 5,528,040 (Lehmann) discloses the use of continuous-wave (c.w.) laser sources for CRDS, in particular laser diode (LD) sources using controlled optical feedback from a reference cavity. Such optical feedback may, however, cause instability in the laser operation. Moreover, locking was turned off during measurements, such that each measurement required re-locking the laser and cavity, which may limit the repetition of rates achievable with the system.

[0006] At least in certain embodiments the present invention is able to provide a system and method for stable and reliable locking of a laser and a ring-down cavity for CRDS, a CRDS system allowing continuous locking of the laser and ring-down cavity over multiple ring-down measurements, a CRDS system allowing high measurement repetition rates, a CRDS system in which locking and sampling light have different frequencies, such that the locking light does not affect measurement values obtained using the sampling light, a CRDS system with reduced shot-to-shot variation in the ring-down constant, a CRDS system with reduced baseline noise, a CRDS system allowing locking of the laser and the ring-down cavity without requiring an external reference cav-

ity, a CRDS system allowing reduced optical feedback from the ring-down cavity to the laser, and a system allowing relatively simply coupling of locking light extending from the CRDS cavity to locking components.

[0007] Thus, the present invention provides a cavity ring-down spectroscopy system comprising:

- (a) a ring-down resonant cavity for holding a sample;
- (b) a light source comprising a laser for generating locking light and sampling light incident on the cavity, the locking light and sampling light being of different polarization and/or of different frequency;
- (c) locking means in optical communication with the cavity for locking the cavity and the light source using locking light reflected by the cavity; and
- (d) detection means in optical communication with the cavity for detecting sampling light extending from the cavity and for allowing determination of ring-down rates indicative of an absorption of the sampling light by the sample.

[0008] The system of the present invention therefore uses distinct sampling and locking light beams. The sampling light is used for performing ring-down measurements, while the locking light is used for frequency-locking a ring-down resonant cavity and a c.w. or other laser. The locking light will in general be maintained continuously on while the sampling light will in general be turned on and off to allow one to obtain ring-down measurements. Continuous locking allows higher measurement repetition rates, and consequently lower noise levels. The locking and sampling light may have different frequencies such that they are effectively decoupled within the cavity, ensuring that the locking light does not affect ring-down measurements performed with the sampling light.

[0009] In a preferred embodiment, the ring-down cavity is a ring-shaped cavity. For a given mode, the ring cavity has distinct resonant frequencies and mirror losses for s-polarized and p-polarized light. If intracavity losses are higher for p-polarized light than for s-polarized light, the sampling light is preferably s-polarized with respect to the cavity, while the locking light is p-polarized. The use of a ring-shaped cavity provides for a desirable separation between the resonant frequencies of a given mode, allowing tuning of each of the sampling and locking light to the same cavity mode without coupling the sampling and locking light. The use of a ring-shaped cavity provides the further advantage of allowing a reduction in optical feedback to the laser.

[0010] At least one of the locking and sampling light is preferably frequency-shifted to ensure that the sampling and locking light are decoupled within the cavity. Preferably, an acousto-optic modulator (AOM) is positioned in the path of the sampling light. The AOM is used to introduce a difference between the frequencies of the sampling and locking light. The AOM is also used as a

switch for turning the sampling light on and off to allow the performance of ring-down measurements.

[0011] In alternative embodiments, the ring-down cavity may be linear or folded. The sampling and locking light can then be tuned to the same transverse mode (e.g. TEM_{00}) corresponding to adjacent longitudinal modes. The frequency-shift between the sampling and locking light is then equal to one free spectral range of the cavity.

[0012] The invention is further illustrated with reference to the accompanying drawings, in which:

Fig. 1-A is a schematic high-level diagram of a preferred system of the present invention,

Fig. 1-B is a schematic diagram of the optical components of the system of Fig. 1-A,

Fig. 1-C illustrates the locking components of the system of Fig. 1-A,

Fig. 2 shows that locking components in an alternative embodiment of the present invention,

Fig. 3 shows an alternative embodiment of the present invention, using a linear ring-down cavity,

Fig. 4 shows scans of the cavity frequency response for p-polarized light and s-polarized light for an exemplary system of the present invention,

Fig. 5 shows a measured plot of the spectral density of the frequency difference fluctuation as a function of frequency difference for the exemplary system characterised by Fig 4, and

Fig. 6 shows a spectrum of water vapour in ambient room air recorded using the exemplary system characterised by Fig 4.

[0013] The following description illustrates the invention by way of example. In the following discussion, unless otherwise stated, polarization directions are to be considered relative to the ring-down resonant cavity of the system. Unless otherwise stated, the term "ring-down" encompass each of decay and build-up (sometimes referred to as "ring-up") of light within the resonant cavity.

[0014] Fig. 1-A is a schematic high-level diagram of a preferred system 20 of the present invention. Optical connections are illustrated by dashed lines, while electrical connections are illustrated by solid lines. System 20 comprises a tunable monochromatic light source 22, a ring-down resonant cavity 24 in optical communication

with light source 22, as well as sampling detection electronics 26, monitoring (diagnostics) electronics 28 and locking electronics 30 all in optical communication with cavity 24. Locking electronics 30 are electrically connected to light source 22 and cavity 24, while monitoring electronics 28 are electrically connected to sampling detection electronics 26.

[0015] Cavity 24 holds a sample of interest in an intracavity light path. Light source 22 generates c.w. input sampling light 32a and locking light 32b incident on cavity 24. Input sampling light 32a has a wavelength within an absorption region of interest of the sample. Input locking light 32b has a predetermined wavelength relationship with sampling light 32a. Sampling light 32a and locking light 32b have different frequencies within cavity 24. Preferably, sampling light 32a is s-polarized while locking light 32b is p-polarized.

[0016] Output light 34a-c extending from cavity 24 is incident on detection electronics 26, monitoring electronics 28, and locking electronics 30, respectively. Output light 34a-c comprises output sampling light 34a incident on sampling detection electronics 26, output monitoring light 34b incident on monitoring electronics 28, and output locking light 34c incident on locking electronics 30. Locking electronics 30 frequency-lock light source 22 and cavity 24, ensuring that the linewidth of intracavity sampling light overlaps the linewidth of a mode of interest of cavity 24. Sampling detection electronics 26 determine ring-down rates, absorption spectra, and species quantities for the sample of interest. Monitoring electronics 28 monitor the intensity and transverse mode structure of intracavity light. The intensity of intracavity light is used to characterize the locking of light source 22 and cavity 24. In one embodiment, measurements of peak intracavity light intensity are used to adjust the frequency separation between locking and sampling light.

[0017] Fig. 1-B illustrates the optics of system 20. Locking electronics 30 are illustrated in Fig. 1-C. For clarity of presentation, various standard elements such as lenses and mirrors used for focusing and directing beams are not described in detail; such elements are well known in the art.

[0018] Referring to Fig. 1-B, light source 22 comprises a continuous wave (c.w.) laser 40 for generating primary light 42, and optical components for generating input sampling light 32a and locking light 32b from primary light 42. Laser 40 preferably has a linewidth on the order of 1 MHz and a useful output power of at least 100 μ W. Laser 40 is preferably, a tunable external-cavity diode laser (ECDL). Other suitable light sources include solid state and dye lasers, as well as C.W. optical parametric oscillators (OPOs). Laser 40, including its controller, is mounted within a Faraday cage (not shown), for shielding laser 40 from stray electromagnetic interference which may otherwise cause instability in the temperature controller of laser 40.

[0019] An optical isolator 44 is positioned in the path

of primary light 42, in optical communication with laser 40. Isolator 44 reduces the optical feedback caused by back reflections into laser 40, and controls the polarization of primary light 42. Isolator 44 is preferably a Faraday isolator including a half-wave plate adjusted to generate p-polarized light. A beam expander 46 and a beam splitter 48 are positioned in sequence in front of isolator 44, so as to receive light extending from isolator 44. Primary light 42 passes through beam expander 46 before being split by beam splitter 48 into sampling light 32a and locking light 32b.

[0020] An acousto-optic modulator (AOM) 54 is positioned in the path of sampling light 32a, between laser 40 and cavity 24. AOM 54 is driven by an RF signal generated by a voltage-controlled oscillator (not shown). AOM 54 is flanked by focusing and mode-matching optics 56a-b. Optics 56a comprise a mode-matching lens for selecting TEM_{00} light for passage to cavity 24, and a focusing lens for collimating sampling light 32a onto AOM 54. Optics 56b comprise lenses for recollimating sampling light 32a after passage through AOM 54. A half-wave plate 57 is positioned in the path of sampling light 32a, for changing its polarization state to s-polarized. In the arrangement shown, AOM 54 is optimized for modulating p-polarized light, and thus half-wave plate 57 is positioned after AOM 54.

[0021] AOM 54 acts as a switch, for modulating (e.g. switching on and off) the intensity of sampling light 32a reaching cavity 24 so as to allow sampling light 32a to ring down within cavity 24.

[0022] AOM 54 may be used to generate pulses or continuous wave step inputs, among others. AOM 54 also acts as a frequency-shifter, for shifting the frequency of sampling light 32a incident on cavity 24 such that s-polarized sampling light 32a is tuned to a desired mode of cavity 24. AOM 54 shifts the frequency of sampling light 32a by an amount equal to the frequency of the RF signal driving AOM 54. Preferably, the first order beam from AOM 54 is directed to cavity 24. An aperture 50 is positioned in the optical path between AOM 54 and cavity 24, to select the first order beam from AOM 54 for passage to cavity 24 while blocking the zero order beam.

[0023] A phase-modulator (PM) 58 is situated in the path of locking light 32b, between laser 40 and cavity 24. PM 58 is preferably an electro-optic resonant phase modulator. In the particular arrangement shown, PM 58 is optimized for modulating s-polarized light, and is thus flanked by half-wave plates 60a-b. Half-wave plates 60a-b control the polarization of light within PM 58 so as to maximize the electro-optic effect within PM 58 for the particular crystal orientation of PM 58. PM 58 introduces locking sidebands into locking light 32b. The sidebands are separated from the central frequency ω_L of locking light 32b by a predetermined frequency separation ω_L larger than the linewidths of laser 40 and the mode of interest of cavity 24. The sideband at $\omega_L + \omega_L$ is phase-shifted by π (180°) with respect to the sideband

at $\omega_L - \omega_L$. A mode-matching lens 62 is situated in the path of locking light 32b, for selecting the TEM_{00} mode for passage to cavity 24.

[0024] A polarizing beam splitter 64 is positioned in the paths of sampling light 32a and locking light 32b, facing cavity 24. Beam splitter 64 combines and directs sampling light 32a and locking light 32b toward cavity 24. Beam splitter 64, isolator 44, and half-wave plates 57, 60a-b act as polarization control elements, selecting s-polarized sampling light 32a and p-polarized locking light 32b for passage to cavity 24.

[0025] Cavity 24 comprises an input mirror 66a, an intermediate mirror 66b, and an output mirror 66c. Mirrors 66a-c define a closed, triangular intracavity light path 68. A gas sample 72 is situated within light path 68, and fills cavity 24. Sample 72 is an optically passive material (i.e. not a gain medium). Input mirror 66a and output mirror 66c are flat moderately-reflective mirrors. Intermediate mirror 66b is a highly-reflective spherical mirror.

20 Intermediate mirror 66b is mounted on a piezoelectric stage 74. Stage 74 is used as a cavity pathlength control element. Stage 74 controls the position of mirror 66b relative to mirrors 66a and 66c, thus controlling the pathlength of cavity 24 and the resonant frequency of any given mode of cavity 24.

[0026] Preferably, the thermal expansion and stress tensor coefficients of cavity 24 are optimized so as to minimize cavity deformations (due to stretching, bowing, bending, etc.) and mechanical resonances. Therefore, it is preferred that the optical elements of cavity 24 are monolithically integrated. Mirrors 66a,c and stage 74 are rigidly attached to a common thermally stabilized block of material (not shown), preferably made of a low-thermal-expansion glass such as mica ceramic. The stable, monolithic attachment of mirrors 66a,c and stage 74 serves to reduce changes in the shape of cavity 24 due to mechanical vibrations and/or temperature variations. Such changes in shape could cause drift in the mode frequencies of cavity 24.

[0027] Input light 32a-b enters cavity 24 through input mirror 66a and generates an intracavity traveling wave along light path 68. Input light 32a-b is not normal to mirror 66a, such that light reflected by mirror 66a is not colinear with light incident on mirror 66a. The finesse of cavity 24 is high enough that the intracavity traveling wave propagates around cavity 24 multiple times. Intracavity locking (p-polarized) and sampling (s-polarized) light encounter different losses within cavity 24, and have different resonant frequencies for a given longitudinal mode of cavity 24. Detection light 34a and monitoring light 34b exit cavity 24 through output mirror 66c.

[0028] A polarizing beam splitter 76 is in optical communication with output mirror 66c. Beam splitter 76 is positioned in an optical path between mirror 66c and a sampling detector 82. Beam splitter 76 directs s-polarized sampling light 34a to sampling detector 82. Detector 82 is preferably a photodiode as described by Harb

et al. in *Phys. Rev. A* 54:4370 (1996), optimized to have a high gain. A polarization separator 78 and a focusing lens 80 are positioned in the optical path between beam splitter 76 and detector 82. Suitable polarization separators include Brewster windows, Glan-Taylor, Glan-Thomson, or Wollaston prisms, or Thomson beamsplitters, among others. Polarization separator 78 selects s-polarized sampling light 34a for transmission to sampling detector 82.

[0029] Ring-down electronics 90 are in electrical communication with sampling detector 82. Ring-down electronics 90 determine ring-down rates and corresponding absorption spectra for sample 72, by analyzing ring-down waveforms detected by sampling detector 82. Ring-down electronics 90 may also determine quantities of trace species of interest in sample 72. The intensity of output sampling light 34a incident on detector 82 is indicative of the intensity of sampling light within cavity 24, and thus of the interaction of sample 72 with intracavity sampling light. Ring-down electronics 90 are conventional. Ring-down electronics 90 may be implemented for example using computer software in an experimental setting, or using dedicated hardware in a process control device in a manufacturing setting.

[0030] Monitoring detectors 86, 88 are in optical communication with mirror 66c. Beam-splitter 76 directs p-polarized monitoring light 34b to monitoring detectors 86, 88, through a beam splitter 92 and a focusing lens 94. Detector 86 is preferably a photodiode as described in the above-referenced article by Harb et al. Detector 86 is used to monitor the power of intracavity light. Detector 86 may be connected through monitoring servo electronics (not shown) to AOM 54, for adjusting the driving frequency of AOM 54 to compensate for changes in the frequency difference between sampling and locking resonances. Such changes may be caused by sample dispersion.

[0031] Detector 88 is preferably a charge-coupled-device array (CCD camera). Detector 88 is used to monitor the transverse mode structure of intracavity light, ideally to ascertain that only TEM_{00} light propagates within cavity 24. Monitoring processing electronics 96 are in electrical communication with detectors 86 and 88, and with ring-down electronics 90. Electronics 96 communicate recorded monitoring data to ring-down electronics 90 for storage with corresponding ring-down data.

[0032] A locking detector 100 is in optical communication with input mirror 66a, and is positioned to capture output locking light 34c extending from input mirror 66a. Locking detector 100 is preferably a photodiode similar to that used for monitoring detector 86, optimized for high bandwidth in order to render the locking sidebands with minimal distortion and noise. A polarization separator 102 and a focusing lens 104 are positioned in an optical path of locking light 34c, between mirror 66a and locking detector 100. Light extending from input mirror 66a consists of a reflected component reflected by input mirror 66a, and of a leakage component transmitted

through input mirror 66a from cavity 24. Polarization separator 102 selects p-polarized locking light 34c for transmission to detector 100.

[0033] Detector 100 is part of locking electronics 30. 5 Fig. 1-C shows a schematic diagram of locking electronics 30. The design of locking electronics 30 is based on the Pound-Drever locking system. For detailed information on Pound-Drever locking see the article by Drever et al. in *Appl. Phys. B* 31:97-105 (1983), herein incorporated by reference.

[0034] Referring to Fig. 1-C, a high-pass filter (HPF) 108 is electrically connected to the output of detector 100. The pass-threshold of HPF 108 is less than the frequency separation ω_f between the central frequency 15 and the locking sidebands of the signal detected by detector 100. A band-pass filter (BPF) 110 is connected to the output of HPF 108. The passband of BPF 110 is centered at ω_f . The output of BPF 110 is connected to an input of a mixer 112. Another input of mixer 111 is connected to a first output of a voltage-controlled oscillator (VCO) 106 capable of generating a driving signal at the frequency ω_f . A second output of VCO 106 is connected to PM 58, for driving PM 58. An output of mixer 112 is connected to a low-pass filter (LPF) 114. The pass-threshold of LPF 114 is less than $2\omega_f$. The output of LPF 20 114 is connected to servo electronics 116, which are in turn connected through an amplifier 118 to stage 74.

[0035] VCO 106 generates a driving signal at ω_f for driving PM 58. PM 58 inserts locking sidebands into locking light 32b at frequencies $\omega_L \pm \omega_f$, where ω_L is the frequency of the central band of locking light 32b. Output locking light 34c is formed by the reflection of locking light 32b and the locking light leaking from cavity 24. The amplitudes of the sidebands of locking light 34c depend on the tuning of each sideband to cavity 24, and consequently on the difference between ω_L and the corresponding resonant frequency of a given mode of interest of cavity 24. For example, if ω_L is higher than a corresponding resonant frequency of cavity 24, the reflected sideband at $\omega_L - \omega_f$ has a lower intensity than the reflected sideband at $\omega_L + \omega_f$.

[0036] Locking light 34c detected by detector 100 includes a frequency component at ω_L and a frequency component at ω_f . Typically, ω_L is on the order of 10^{14} 45 Hz, while ω_f is on the order of 10^7 Hz. Detector 100 is not fast-enough to time-resolve the ω_L component of locking light 34c. Consequently, the electric signal generated by detector 100 includes a component centered at zero frequency, corresponding to the ω_L frequency component of locking light 34c. Detector 100 can time-resolve the ω_f component of locking light 34c, and thus the generated electric signal includes a component centered at ω_f . HPF 108 and BPF 110 select the ω_f frequency component for passage to mixer 112.

[0037] Mixer 112 mixes the signals received from VCO 106 and detector 100 to generate an error signal centered at zero frequency. Mixer 112 effectively differences the sideband signals to generate the error signal.

The sign of the error signal indicates whether ω_L is higher or lower than the corresponding resonant frequency of cavity 24. LPF 114 eliminates higher-harmonics (e.g. $2\omega_L$) from the signal generated by mixer 112, selecting the zero-frequency error signal for passage to servo electronics 116. Servo electronics 116 translate the error signal into an appropriate motion of stage 74, for adjusting the pathlength and thus resonant frequency of interest of cavity 24 to ensure that a desired mode of cavity 24 is tuned to ω_L .

[0038] Locking light 32b is continuously turned on during the operation of system 20, for ensuring continuous locking of cavity 24 and light source 22. Sampling light 32a is selectively turned off after sufficient accumulation of light within cavity 24 during periods of c.w. operation, to allow measurements of the ring-down of sampling light 32a within cavity 24. Maintaining locking light 32b turned on while performing multiple ring-down measurements allows relatively high repetition rates (e.g. kHz to tens of kHz) for system 20.

[0039] Fig. 2 is a simplified diagram illustrating an alternative system 20' according to the present invention. Locking electronics 30' control the frequency of light emitted by a laser 40', so as to tune the central band of locking light 32b to cavity 24. The output of mixer 112 is connected through LPF 114, servo electronics 116', and an amplifier 118' to a frequency controller of laser 40'. If ω_L is higher than the resonant frequency of interest of cavity 24, servo electronics 116' control laser 40' to reduce its emission frequency so as to tune ω_L to the desired mode of cavity 24. Similarly, ω_L is increased if it is lower than desired.

[0040] Fig. 3 is a simplified diagram illustrating another alternative system 20" of the present invention. System 20" includes a linear ring-down cavity 24". A polarizing beam splitter 65 and a quarter-wave plate 67 are positioned in sequence between PBS 64 and cavity 24", in the optical path of the sampling and locking light. PBS 65 directs incident locking and sampling light toward cavity 24", while directing reflected locking light toward detector 100. The frequencies of the sampling and locking light are tuned to the same transverse mode (e.g. TEM₀₀) but different (e.g. adjacent) longitudinal modes of cavity 24". The frequency difference between the sampling and locking light is equal to one free spectral range (FSR) of cavity 24". For typical FSR values of hundreds of MHz, a conventional AOM is well suited for introducing the desired frequency difference between the sampling and locking light.

[0041] Baseline noise in CRDS is caused in large part by concurrent excitation and beating of multiple modes within the ring-down cavity (RDC), or by consecutive excitation of different modes. Different transverse/longitudinal modes have different resonant frequencies, and ring-down at different rates in an empty cavity. Ensuring that a single mode is excited within the ring-down cavity allows a significant improvement in system signal-to-noise ratios.

[0042] Because frequency fluctuations in the laser source appear identically in both sampling and locking light, if a locking light mode is locked to the cavity, then a corresponding sampling light mode is also locked to the cavity. The AOM used for switching the sampling light on and off provides the appropriate frequency shift to the sampling light, so that it can couple into the same transverse mode as the locking light.

[0043] Using a ring-shaped ring-down cavity in a system of the present invention provides a number of advantages. The use of a ring-shaped cavity provides for the desired separation between the resonant frequencies for sampling and locking light of different polarizations, allows a simplification in the optics used for coupling locking light into and out of the cavity, and allows a reduction in optical feedback to the laser.

[0044] It is generally well-known from the physics of optical interfaces that non-normal incidence of linearly polarized light on a dielectric interface will usually result in different responses for p-polarized light (PPL) and s-polarized light (SPL). Thus, an optical ring resonator constructed with a geometry using non-normal incidence reflectors (e.g., an isosceles triangle) will actually consist of two superimposed nondegenerate Fabry-Perot cavities: a p-polarization cavity (PPC), and an s-polarization cavity (SPC).

[0045] Non-normal incidence dielectric mirrors typically have a lower reflectivity for PPL than they do for SPL. Consequently, a ring resonator will usually consist of a reduced finesse PPC, and a higher finesse SPC. The ring resonator PPC and SPC transverse mode structures are identical, because they are determined only by geometrical considerations. The unequal phase shifts accrued during non-normal reflection of PPL and SPL will result in a fixed frequency difference between the PPC and SPC frequency responses. PPC and SPC free spectral ranges differ by the contribution of the derivative of the mirror phase shift, which was estimated to be a few tens of kHz.

[0046] Because orthogonal polarizations are easily separated with polarizing optics, the simultaneous use of PPL and SPL provides a straightforward solution for separating the cavity locking problem from the actual ring-down measurement. The use of the lower finesse PPC for locking improves the feedback locking signal, and relaxes requirements on servo bandwidth, feedback gain, and differential sensitivity to the frequency difference between the laser and cavity. Absolute wavelength accuracy at each point is determined by the PPC finesse and locking servo quality, rather than by laser linewidth or cavity mode sweep range. The improved frequency stability enables a substantial increase in the resolution achievable with such a system.

[0047] The use of a ring cavity also allows a significant simplification in the optics used to couple locking light into locking components. In a system using a linear or folded cavity, the optical signal extending from the cavity input is colinear with the light beam incident on the cavity

input. As a result, optical components such as half-wave plates, analyzers, and beam-splitters may be needed to selectively couple the locking light reflected by the cavity input into the locking electronics. Furthermore, ring cavities do not reflect light directly back into the light source, so that optical feedback problems are greatly reduced.

[0048] The following example is intended to illustrate a particular implementation of the invention, and should not be construed to limit the invention.

Example

[0049] A system of the present invention was used to generate spectra of water vapor in ambient air. The laser was a commercial ECDL (New Focus: 6226-H032), tunable from 833.2 nm to 862.5 nm. The laser output varied from about 9 to 13 nW for 60 mA drive current, over the entire tuning range. A 25-35 dB Faraday isolator (New Focus: 5568) was used to reduce residual back reflections into the laser. The input and output mirrors of the RDC (CVI:TLM2-800-45S-1037) had 99.93% reflectivity for SPL at 833 nm, and 99.3% reflectivity for PPL at 833 nm. The intermediate RDC mirror (REO: run C628, 7.75 mm, 840-880 nm, 1 degree wedge) had a 99.95% reflectivity at 833 nm for both polarizations.

[0050] One quarter of the primary light generated by the laser was used for locking and three quarters for sampling. The input sampling light was focused to a spot size of 60 μ m at the center of the AOM (Brimrose: GPM-400-100-960), and recollimated using 5 cm focal length lenses. The AOM was driven by a VCO with a frequency range of 300 to 535 MHz. The AOM imparted a frequency shift of 320 MHz to the input sampling light. The lenses used for TEM₀₀ mode-matching of the sampling and locking light had a focal length of 127 cm. The phase modulator was a 58.5 MHz electro-optic resonant phase modulator (New Focus:4001). The fraction of laser light coupled into the TEM₀₀ mode of the cavity was ~90% for PPL, and ~85% for SPL.

[0051] The VCO driving the phase-modulator was set to 58.5 MHz. A Glan-Taylor prism was used to select PPL for passage to the locking detector. The bandpass of the locking bandpass filter was centered at 60 MHz. The servo electronics consisted of a proportional integral (PI) controller coupled to a passive notch filter centered at 52 kHz (48 dB maximum attenuation, 3 dB bandwidth of 2 kHz). The notch filter was used to suppress oscillation of the piezoelectric stage at its resonant frequency of 52 kHz. The use of the notch filter improved the stability of the system and reduced its sensitivity to external perturbations such as acoustic noise. The gain of the servo electronics and associated amplifier was 60 dB, with a 3 dB bandwidth of 455 Hz.

[0052] The locking and monitoring photodiodes were optimized for a bandwidth of 90 MHz. Shot-noise-limited sideband signals were readily attainable for optical powers of hundreds of μ W at the detectors. The sampling photodiode was optimized for gain, at a bandwidth of 25

MHz. The output sampling light power was on the order of μ W, and the detected ring-down decay constants were on the order of hundreds of ns or higher. The monitoring photodiode received 90% of the monitoring light, while the monitoring CCD camera received the remaining 10%.

[0053] A Glan-Thomson prism was used to select SPL for passage to the sampling detector. After detection by the sampling detector, the sampling signals were amplified using a low-noise, highspeed amplifier (Stanford Research Systems:SR445), and digitized using a 10-bit, 1 GHz oscilloscope (Tektronix:11402). The decay waveforms were fitted using the Levenberg-Marquardt algorithm with the initial guess provided by a linear least squares fit of the signal logarithm, as described by Naus et al. in *ILS-XII 12th Interdisciplinary Laser Science Conference*, ed. A. P. Society (OSA, Rochester, New York, 1996), p. 122. All scans were performed in air at atmospheric pressure, with a water partial pressure of 12 torr.

[0054] Fig. 4 shows scans of the cavity frequency response for PPL and SPL. The zero frequency is arbitrary. The resonance frequencies for SPL and PPL were observed to be separated by 282 MHz. The resonance frequency separation changed by several tens of kHz when the cavity modes overlapped an absorption line of water. The driving frequency for the AOM was accordingly adjusted using intensity information from the monitoring electronics, to maintain an appropriate frequency separation between the locking and sampling light. For small sample quantities (e.g. for trace species detection), the frequency separation resulting from dispersion by the sample is generally small or negligible.

[0055] The discriminator slope of the error signal generated by the mixer, which converts error signal voltage into frequency difference, was used to determine fluctuations in the frequency difference between the laser frequency and the cavity resonance frequency. Fig. 5 shows a measured plot of the spectral density of the frequency difference fluctuation (noise as a function of frequency difference between the laser and cavity). The total jitter between the laser and cavity resonances was 1.1 MHz, slightly less than the free-running laser linewidth. The locking components reduced noise at low frequencies (<20 kHz). Faster servo electronics may generally be used for reducing higher frequency components of the noise.

[0056] Fig. 6 shows a spectrum of water vapour in ambient room air recorded using the system described above, and a corresponding spectrum obtained from the HITRAN96 database. The resolution of the recorded spectrum was 0.002 nm, except about the absorption peaks where the resolution was increased to 0.001 nm. Ten averaged decay waveforms (ADW) were recorded at each wavelength. Each ADW was determined by averaging 256 shots. The RMS baseline noise was 5×10^{-9} cm⁻¹, and the nominal sensitivity was 5ppm. The recorded spectrum compares favourably with the HITRAN96

spectrum in absolute frequency, line strength, and linewidth.

[0057] The above embodiments may be altered in many ways. For example, a wide variety of wavelength ranges may be used, and the presence of a variety of species may be determined. Various locking schemes and electronics may be used.

Claims

1. A cavity ring-down spectroscopy system comprising:

- (a) a ring-down resonant cavity for holding a sample;
- (b) a light source comprising a laser for generating locking light and sampling light incident on the cavity, the locking light and sampling light being of different polarization and/or different frequencies;
- (c) locking means in optical communication with the cavity for locking the cavity and the light source using locking light reflected by the cavity; and
- (d) detection means in optical communication with the cavity for detecting sampling light extending from the cavity and for allowing determination of ring-down rates indicative of an absorption of the sampling light by the sample.

2. A system according to claim 1, wherein one of the locking light and the sampling light is s-polarized within the cavity, and the other of the locking light and the sampling light is p-polarized within the cavity.

3. A system according to claim 1 or 2, wherein the locking means is in electrical communication with at least one of the light source and the cavity, receives locking light reflected by the cavity, and locks the cavity and the light source using the locking light reflected by the cavity.

4. A system according to any preceding claim, wherein the light source further comprises polarization control optics in an optical path between the laser and the cavity, for selecting a p-polarization for the locking light, and an s-polarization for the sampling light.

5. A system according to any preceding claim, wherein the light source further comprises frequency-shifting optics in an optical path between the laser and the cavity, for frequency-shifting at least one of the locking light and the sampling light to tune said at least one of the locking and the sampling light to a desired mode of the cavity.

6. A system according to claim 5, wherein the frequency-shifting optics comprises an acousto-optic modulator positioned in an optical path of the sampling light between the laser and the cavity, for frequency-shifting the sampling light to tune the sampling light to the cavity, and for switching the sampling light to allow the sampling light to ring down within the cavity.

10 7. A system according to any preceding claim, wherein the cavity has a cavity pathlength control element in electrical communication with the locking means, for controlling an optical pathlength of the cavity to lock the cavity to the light source.

8. A system according to any preceding claim, wherein the light source further comprises a phase modulator in an optical path of the locking light between the laser and the cavity; and wherein the locking means comprises:

- a) a locking detector in optical communication with the cavity, for detecting the locking light reflected by the cavity;
- b) processing electronics in electrical communication with the locking detector, for generating an error signal indicative of a difference between a central frequency of the locking light and a resonant frequency of the cavity;
- c) an oscillator in electrical communication with the phase modulator, for driving the phase modulator to insert a first and a second sideband in the locking light, the first sideband being at a higher frequency and the second sideband being at a lower frequency than the central frequency; and
- d) a mixer in electrical communication with the locking detector and the oscillator, for differentiating electrical signals corresponding to the first and second sidebands to generate the error signal.

9. A system according to any preceding claim in which:

- a) the laser is a continuous-wave tunable laser, in optical communication with an input to the cavity, for generating a primary light beam;
- b) the system additionally comprises beam splitting optics in an optical path between the laser and the input, for splitting the primary beam into the locking light and the sampling light incident on the input, the locking light being p-polarized and the sampling light being s-polarized within the cavity;
- c) the locking means generates an electrical feedback error signal from a reflection of the locking light from the input, and locks the laser

and the cavity using the error signal; and
d) a switch is positioned between the beam
splitting optics and the input, in an optical path
of the sampling beam, for switching the sam-
pling beam to allow the sampling beam to ring
down within the cavity multiple times while the
laser and the cavity are locked. 5

10. A system according to any preceding claims, in
which the detection means detects a plurality of
ring-downs of sampling light extending from the
cavity while the cavity and the light source are
locked. 10

11. A system according to any preceding claim, in
which the detection means detects multiple ring-
downs of the sampling light extending from the cav-
ity while the cavity and the light source are locked;
the system additionally comprising ring-down
means in electrical communication with the detec-
tion means, for determining ring-down rates for the
ring-downs. 15 20

12. A system according to any preceding claim, where-
in the cavity is a linear cavity. 25

13. A system according to claim 12, wherein a frequen-
cy separation between the sampling light and the
locking light is substantially equal to one free spec-
tral range of the cavity. 30

14. A method of locking a laser and a cavity for ring-
down spectroscopy, comprising the steps of:

a) using the laser to generate sampling light
and locking light incident on the cavity, wherein
the sampling light and the locking light have dif-
ferent frequencies; 35

b) using the locking light to lock the cavity and
the laser; and 40

c) obtaining a plurality of measurements of a
ring-down of the sampling light while maintain-
ing the cavity and the laser in a locked state,
for generating an absorption spectrum of a
sample situated within the cavity. 45

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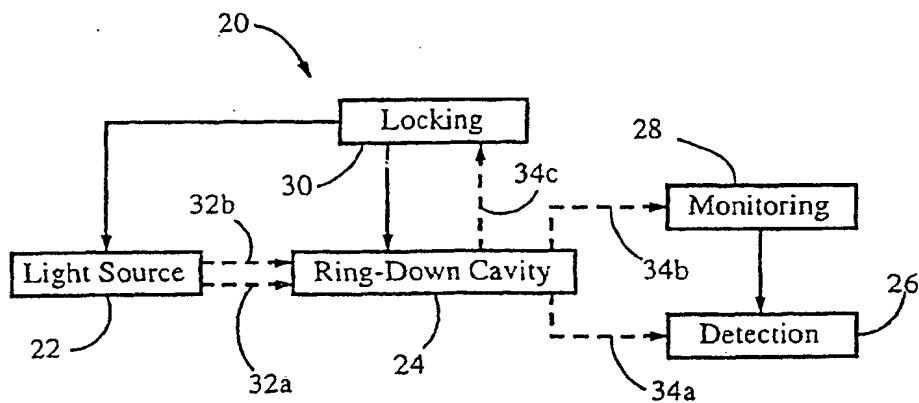


FIG. 1-A

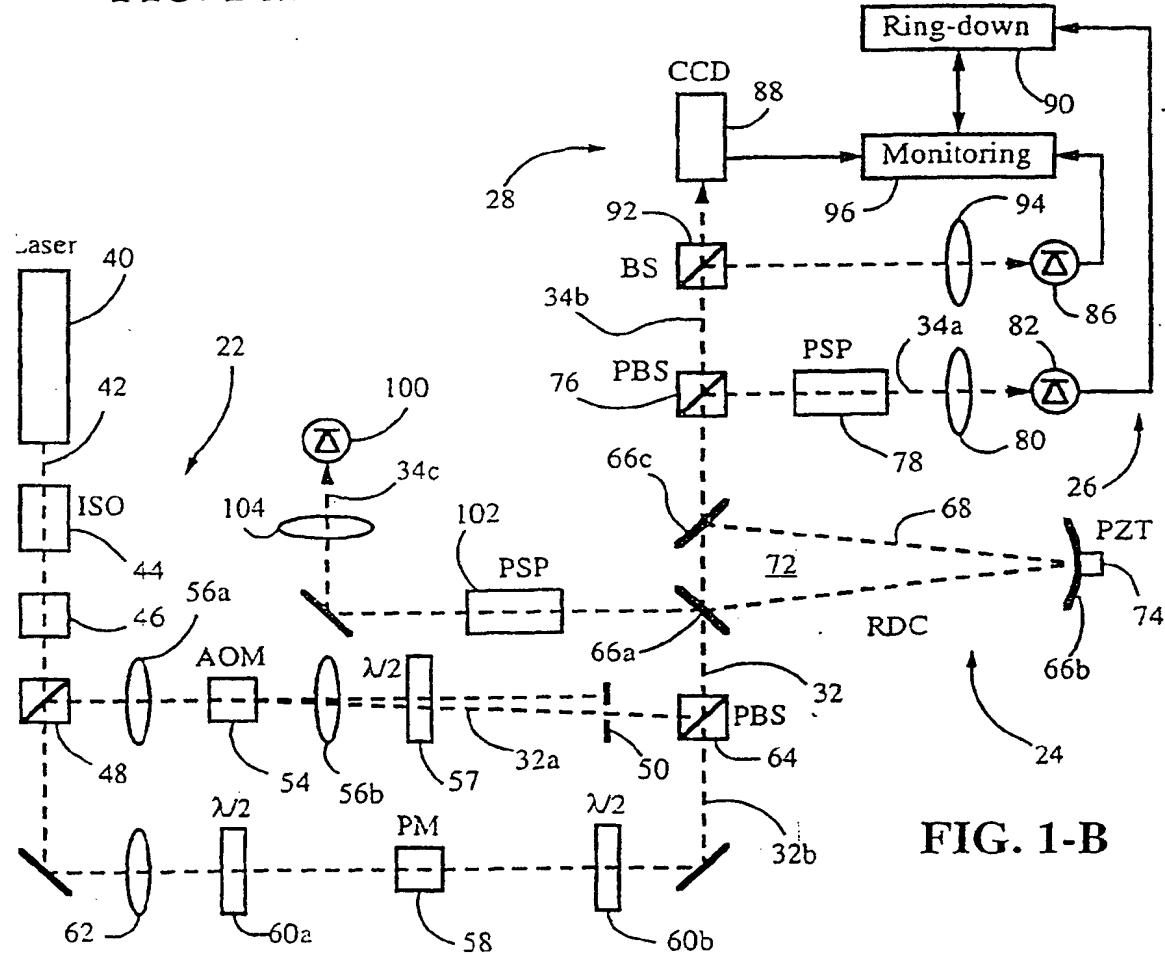


FIG. 1-B

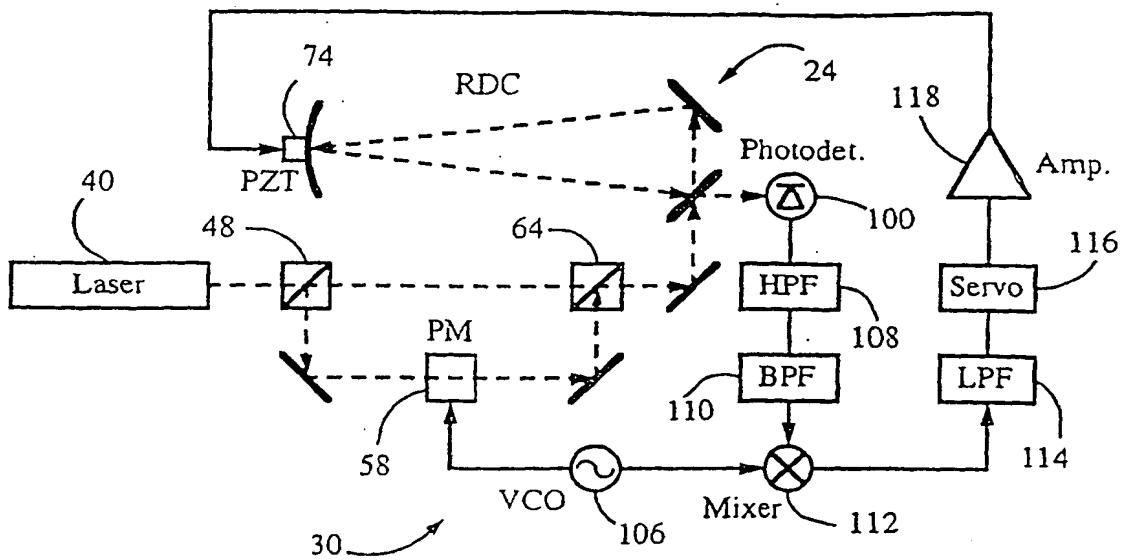


FIG. 1-C

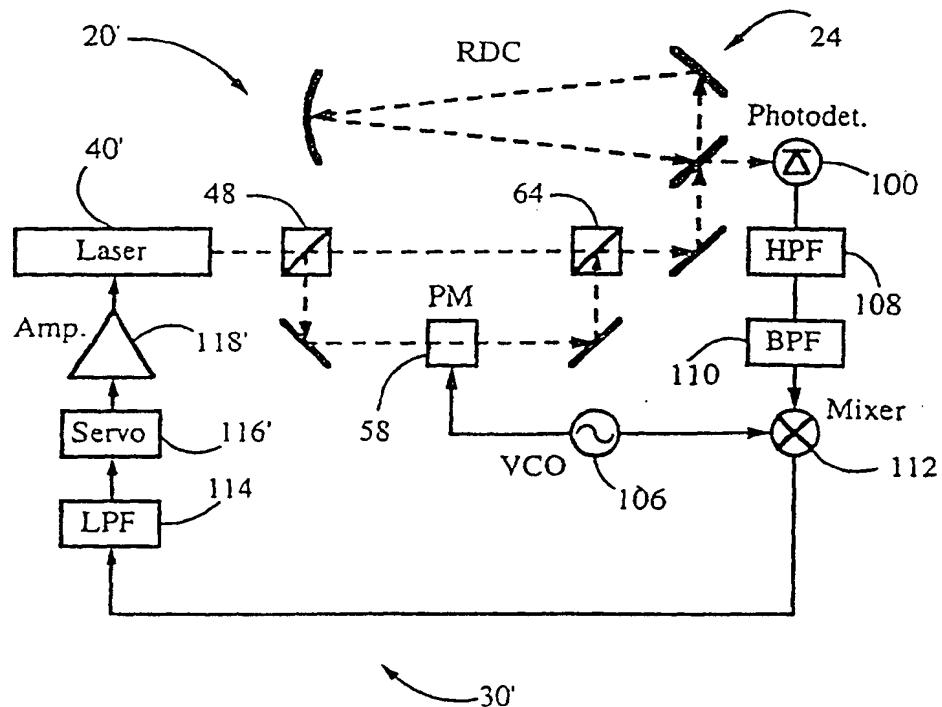


FIG. 2

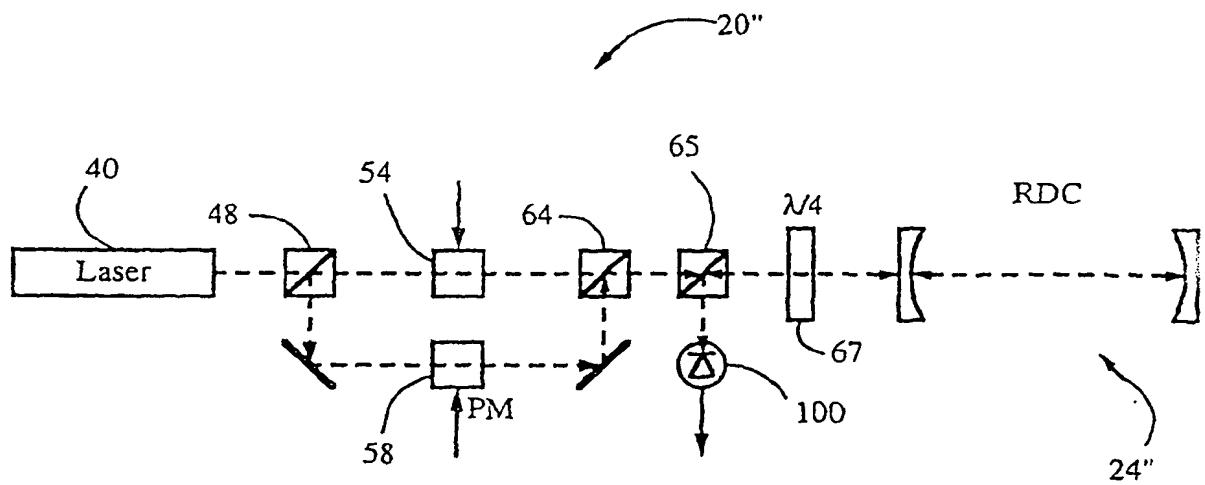


FIG. 3

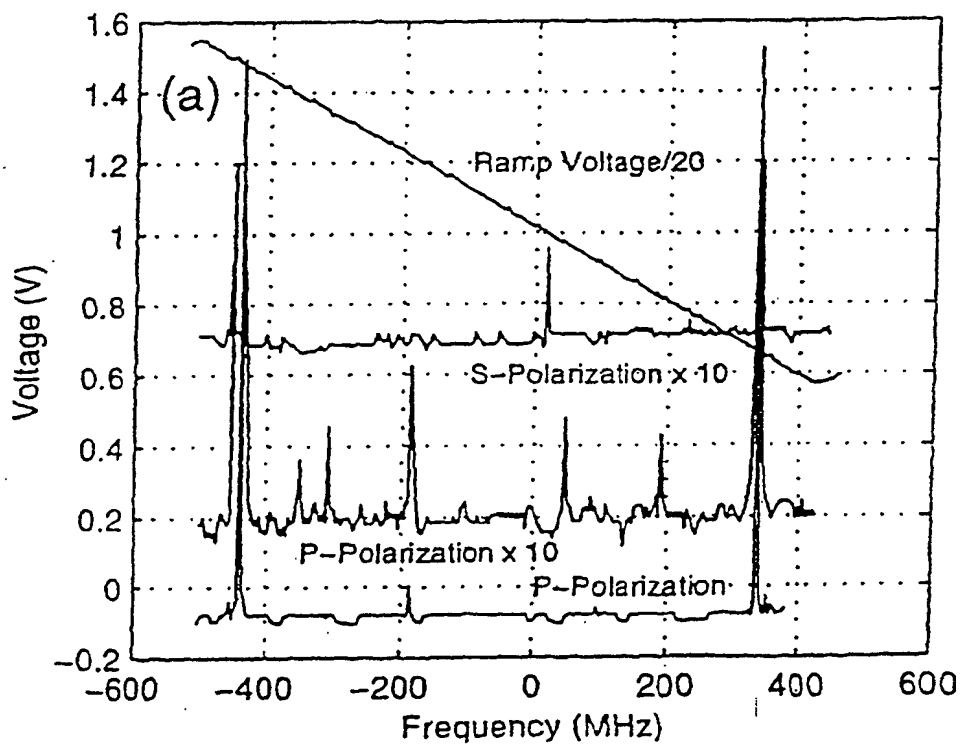


FIG. 4

FIG. 5

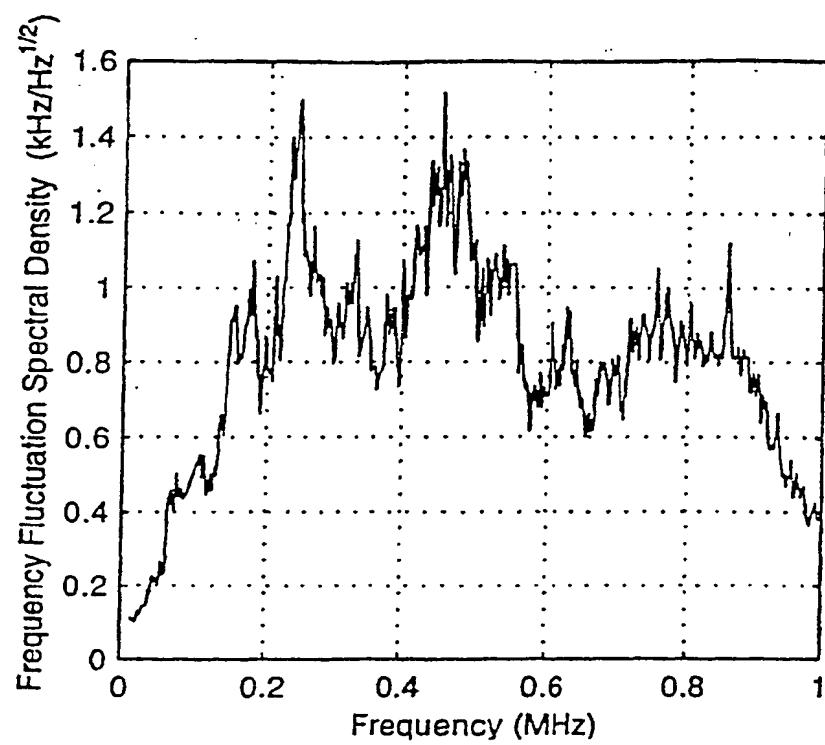
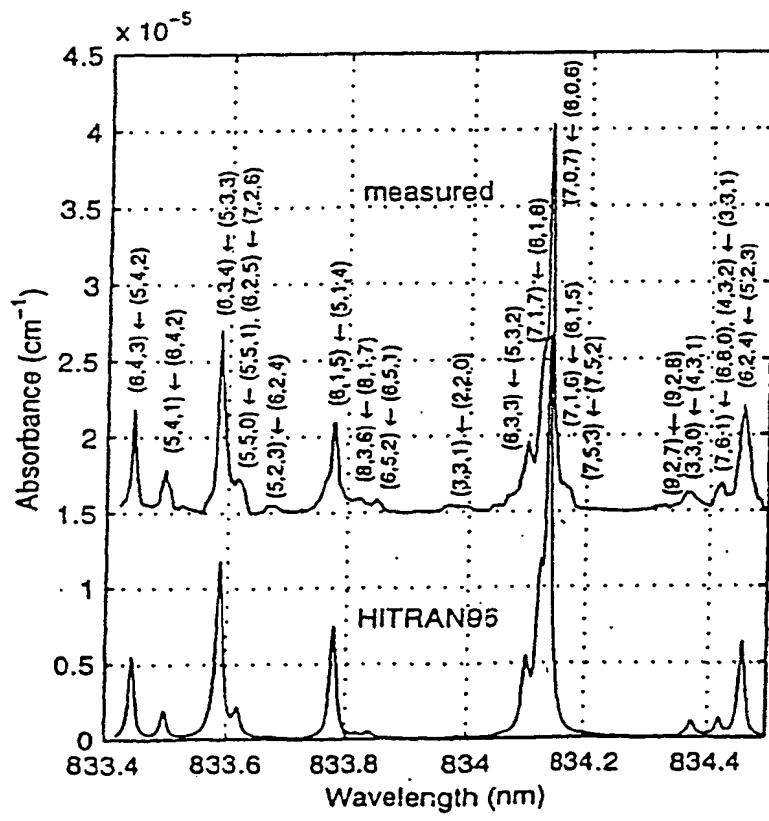


FIG. 6





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EUROPEAN SEARCH REPORT

Application Number
EP 00 30 0294

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Place of search	Date of completion of the search	Examiner	
MUNICH	27 June 2000	Mason, W	
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P : intermediate document	& : member of the same patent family, corresponding document		



EUROPEAN SEARCH REPORT

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EP 00 30 0294

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			TECHNICAL FIELDS SEARCHED (Int.Cl.7)						
<p>The present search report has been drawn up for all claims</p> <table border="1"> <tr> <td>Place of search</td> <td>Date of completion of the search</td> <td>Examiner</td> </tr> <tr> <td>MUNICH</td> <td>27 June 2000</td> <td>Mason, W</td> </tr> </table> <p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document</p>				Place of search	Date of completion of the search	Examiner	MUNICH	27 June 2000	Mason, W
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MUNICH	27 June 2000	Mason, W							

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